The Interaction of Water and Aerosols in the Marine Boundary Layer: A Study of Selected Processes Impacting Radiative Transfer and Cloudiness

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LONG-TERM GOALS

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

OBJECTIVES

For the current reporting period, our efforts have centered on the continued analysis of the data we gathered during the CARMA-IV field campaign, commencement of a retrospective analysis of data from the CARMA-II and CARMA-III campaigns, and a preliminary assessment of data from the VOCALS campaign. Our objectives for these analyses have changed somewhat from those in our original proposal in light of our findings to date. We summarize them as follows.

- Using the same receptor modeling analysis for source attribution that we employed for the size-resolved aerosol hygroscopicity (Hegg et al, 2008), develop a similar source attribution for the CCN concentration in the marine atmosphere off the California coast.

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14. ABSTRACT

The overarching, log term goal of the study is to explore the profound effect of aerosol-water interaction both on radiation propagation in, and the thermodynamic structure of, the marine boundary layer. Specific goals are: 1) compile a climatology of aerosol hygroscopicity for use in the NAAPS and COAMPS models, and, further, to develop a model parameterization of hygroscopicity based on aerosol size and composition for such models, 2) explore the relative impacts of cross-inversion mixing and sub-cloud aerosol on cloud thickness and cloud base height, 3) quantify and parameterize the impact of precipitation scavenging on below cloud radiative transfer and cloud liquid water path. The sampling platform utilized is the CIRPAS Twin Otter research aircraft and the venue is the littoral environment off the California coast, representative of areas with high shipping densities.

15. SUBJECT TERMS

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- Perform the same sort of CCN source attribution done in CARMA-IV on data from CARMA-II and CARMA-III to assess inter-annual variability in the CCN sources.

- Assess the importance of elemental carbon (EC) particles as CCN using data from both the CARMA studies and, if feasible, VOCALS.

- Develop a climatology of aerosol optical properties based on data from the CARMA field studies plus data from ACE-2, ACE-Asia and RED field campaigns.

APPROACH

The source attribution of CCN requires the measurement of both the aerosol chemical composition and the CCN activity. The first is measured via chemical analysis of filter samples by means of a variety of analytical techniques (Gao et al, 2003). The second is done with the DMT Inc. CNN-100 CCN spectrometer that employs the design described by Roberts and Nenes (2005). It measured cumulative CCN number concentrations at nominal supersaturations of 0.2, 0.3, 0.5, 0.7 and 1.0 %. The techniques described above were deployed from an airborne platform, the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter research aircraft. This platform, and its associated facility instruments, has been described in a number of publications (e.g., Wang et al, 2002; Schmid et al, 2003; Hegg et al, 2005).Because source profiles for aerosol in this area are not known, we have not employed fully deterministic source assessment (i.e., a chemical mass balance model) but rather alternative ANOVA techniques, specifically, the EPA PMF 1.1 and UNMIX 2.3 models. These are operational, regulatory, receptor models used precisely to deconvolute aerosol sources when good source profiles are not available. Additionally, we have tested this approach with a data set from the arctic to ensure its feasibility (Hegg et al, 2009b).

The second objective is addressed by the same approach as the first, but now applied to other data sets (e.g., CARMA-II, CARMA-III, ACE-Asia, etc.)

The methodology for achieving the third objective involves the use of two instrument new to our program. The first of these is the annular geometry CCN spectrometer manufactured by DMT Inc. to which we alluded above. This will yield a continuous record of the CCN concentration at five supersaturations with a time resolution of about 10 minutes. This data will be compared with the concentration of EC bearing particles measured by the SP2 instrument manufactured by DMT Inc. and recently evaluated and described by Moteki and Kondo (2007). The comparison will yield the fraction of the CCN number concentration at each supersaturation that contain EC.

The final objective, the development of a climatology of aerosol optical properties is addressed in a straightforward manner using standard statistical techniques to derive the distributions of the various optical properties over the various field campaigns for which we have data.

WORK COMPLETED

The analysis of the data from the CARMA-IV campaign has been partially completed, with results pertinent to our first and fourth objectives listed above in hand. The third objective has been addressed but not yet fully completed. As mentioned in the last annual report, problems with the analysis of the SP2 data have hindered the analysis. However, much of this has been resolved and some preliminary analysis completed The second objective, the analysis of the multiple linkages between cloud optical

depth and albedo, and the proximate parameters that control them, has required some adjustments on our part. The main thrust of our analysis was to have been the examination of the impact of inhomogeneous mixing on cloud albedo as per Burnet and Brenguier (2006). However, our analysis of all CARMA data has yielded only one possible instance of such mixing. Hence, it is our current view that this process is so rare that it has no significance, at least for our venue. Consequently, we will be re-directing our analysis to address the issue of the relative impact of conventional mixing and aerosols on stratocumulus albedo.

RESULTS

The source attribution of the aerosol mass derived from the receptor modeling analysis (specifically UNMIX 2.3) has been reported in Hegg et al (2008) and will not be discussed here. Based on this source attribution of aerosol mass, a source attribution of the concurrently measured CCN activity could be made with standard regression analysis. The results of this analysis, in terms of the cumulative CCN spectra for each of the aerosol types are shown in Figure 1. The slope of the CCN activation spectrum is clearly steepest for the pollution-derived aerosol, as expected. Indeed, all of the slopes are as expected, supporting the validity of the source attribution.

To assess which aerosol types the clouds in the CARMA operational area actually form on, an additional step is necessary. The typical supersaturation in the clouds must be assessed and the concentration of CCN at this supersaturation used in the source attribution. Such an assessment has been made (Hegg et al, 2009a) and 0.3% supersaturation found to be the typical value, in agreement with numerous other studies on marine stratocumulus. Hence, we use the CCN concentration at 0.3% supersaturation in our source analysis for the entire data set. The partitioning of the CCN active at 0.3% supersaturation for each of the samples is shown in Figure 2. As expected, the marine source is largest but there is substantial variation between samples. CCN samples 4 and 6 are dominated by biomass burning aerosol as was the mass, while pollution is the most prominent source in samples 13 and 17-21, associated with the transport of pollution from the South coast Air Basin. Averaged over all of the samples, the marine source contributes 51% of the CCN number concentration, the pollution 30% and the biomass burning 19%. As expected, this differs somewhat from the mass attribution.

IMPACT/APPLICATIONS

The results of the CCN source analysis from CARMA-IV support the conceptual picture of a mixed source aerosol in the littoral area off the California coast. The ability of the receptor model to differentiate between natural CCN and anthropogenic suggests it will be a valuable tool in determining the impact of anthropogenic CCN on indirect climate forcing.

TRANSITIONS

None.

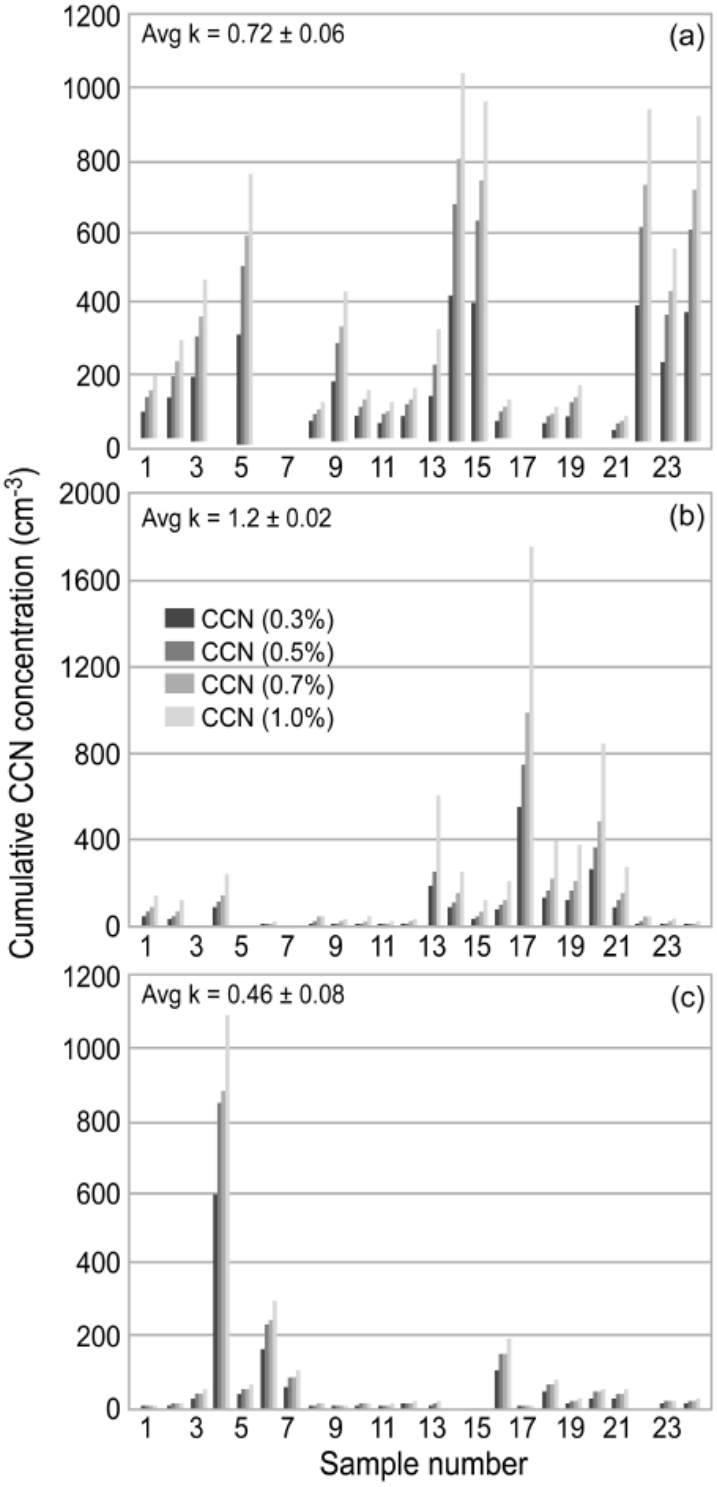

Figure 1. Bar graph of cumulative CCN spectra in terms of number of active CCN at successively higher supersaturation for each of the samples. Panel (a) is for the marine component, panel (b) for the pollution and panel (c) for the biomass smoke. It is clear that the pollution-derived aerosol have the most rapidly increasing CCN activity as a function of increasing supersaturation.

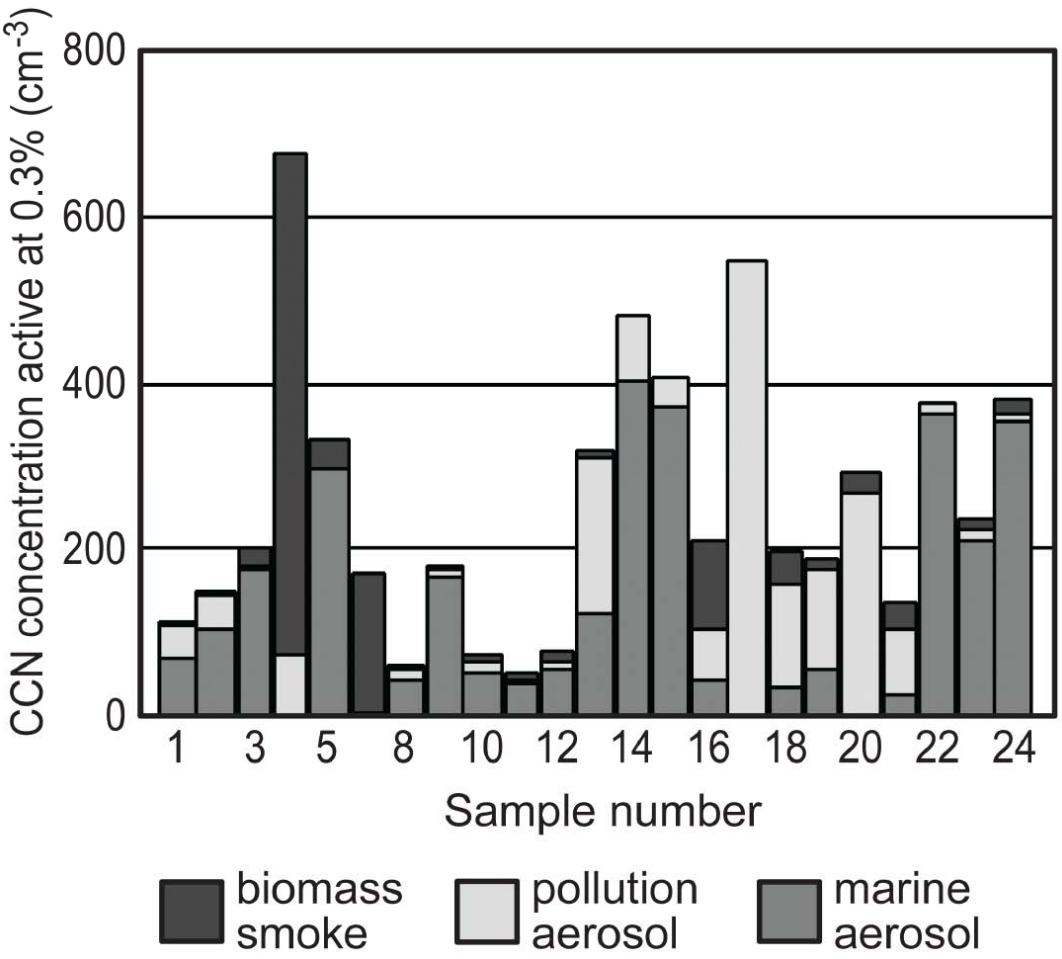

Figure 2. Bar graph showing the source contributions of the three main sources of CCN to each sample taken during the CARMA-IV study.
In most instances, marine aerosol dominated the samples.

RELATED PROJECTS

These measurements are highly relevant to determination of CCN spectra and thus of the microphysics of MBL clouds. Furthermore, numerical transport models now incorporate aerosol sources and can predict the amount of aerosol from the incorporated sources present at any particular point in the model domain. The ability to differentiate between CCN from different sources should permit a source-specific characterization of the CCN activity to use in such transport models, thus improving their prognostic power.

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PUBLICATIONS

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PATENTS

None.